Giant strain and induced ferroelectricity in smorphous $BaTiO_3$ films and multiferroic heterostructure under poling

P. Mirzadeh Vaghefi[1], A. Baghizadeh[2], A.A.C.S. Lourenço[1], V.S. Amaral[1], A.L. Kholkin[1,3]

[1]*Department of Physics & CICECO-Aveiro Institute of Materials, University of Aveiro, 3810-193 Aveiro, Portugal*
e-mail: kholkin@ua.pt

[2]*Department of Materials and Ceramics Engineering & CICECO - Aveiro Institute of Materials, University of Aveiro, 3810-193 Aveiro, Portugal*

[3]*School of Natural Sciences and Mathematics, Ural Federal University, 620000 Ekaterinburg, Russia*

Amongst the benefits of nanotechnology in science and industrial applications, it offers novel high resolution nanopatterning techniques that are useful to overcome the restrictions of conventional lithography [1,2]. With that purpose, several lithographic methods have been developed in past decades, including those based on scanning probe microscopy (SPM) [3]. These methods offer important advantages, such as a few nanometers resolution and high versatility. In the recent years, much attention has been paid to the ferroelectric lithography that relies on the polarization reversal by the tip of piezoresponse force microscope (PFM) exposing to polarization sensitive media [4]. These methods use local polarization reversal accompanied by the reversible strain that very often does not exceed 1% even under applying of a very high electric field [5,6]. Morphological changes due to the application of an external electrical bias, i.e. electric poling treatment have been previously observed and attributed to the interaction between conductive probe and the trapped charges injected into the film during poling process in polymers [5] and PZT/LSMO [6] thin films. In oxide ferroelectrics, these effects have not been seen so far.

In this work, we report the giant surface modification of a 5.6 nm thick $BaTiO_3$ film grown on Si (100) substrate under poling by conductive tip of a scanning probe microscope. The surface is locally elevated by about 9 nm under -20 V applied voltage during scanning, resulting in the maximum strain of 160%. The threshold voltage for the surface modification is about 12 V. The modified topography is stable enough with time and slowly decays after poling with the rate ~0.02 nm/min. Also, strong vertical piezoresponse after poling is observed.

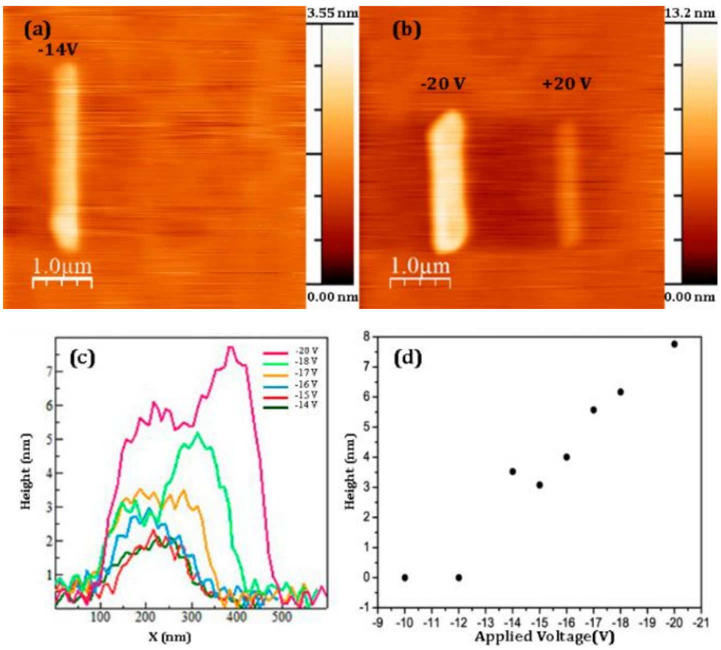

Figure 1. Topography of $BaTiO_3$ thin film after application of (a) ±14 V and (b) ±20 V during scanning of the area 0.2x2 μm^2. (c) Comparison of the topography cross-sections of the areas poled with different voltages. (d) Average height of the poled areas vs. applied negative voltage.

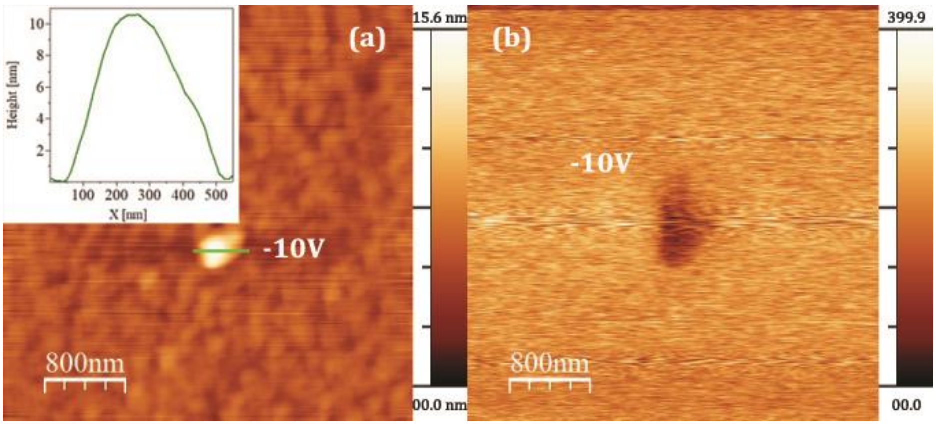

Figure 2. (a) Topographic AFM change of the LBMBT surface after poling of -10V and (b) piezoresponse of the poled area.

Combined measurements by SPM and piezoresponse force microscopy (PFM) prove that the poled material develops high ferroelectric polarization that cannot be switched back even under oppositely oriented electric field. The topography modification is hypothesized to be due to a strong Joule heating and concomitant interface reaction between underlying Si and $BaTiO_3$. Top layer is supposed to become ferroelectric as a result of local crystallization of amorphous $BaTiO_3$.

Later, we extended the patterning method to a multiferroic heterostructure of magnetic layers ($La_{0.9}Ba_{0.1}MnO_3$) covering 30 nm $BaTiO_3$ layer [7]. Coupling of polarization, in-plane strain and magnetic properties in a heterostructures can be used to engineer the symmetry of thin films and heterostructures and be applied in ferromagnetic shape memory alloys [8-10]. The surface of heterostructure is locally elevated by 10.5 nm under -10 V applied voltage during scanning, resulting in 135% strain. Asymmetrical polarization was observed regarding the sign of applied voltage. This work illustrates a novel approach of nanopatterning of ferroelectric films and creation of ferroelectric nanodomains by means of PFM technique and opens up new ways to create nanoscale ferroelectric structures to be used in ferroelectric memory devices.

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